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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(21) International Application Number: PCT/US97/22892 (22) International Filing Date: 15 December 1997 (15.12.97) (30) Priority Data: 08/775,660 31 December 1996 (31.12.96) US (71) Applicant (for all designated States except US): THE DOW CHEMICAL COMPANY [US/US]; 2030 Dow Center, Midland, MI 48674 (US). (72) Inventors; and (75) Inventors/Applicants (for US only): WANG, David, S. [US/US]; 214 Raintree Lane, Lake Jackson, TX 77566 (US). ANDERSON, Kenneth, W. [US/US]; 54 Banyan Court, Lake Jackson, Texas 77566 (US). WHITE, Jerry, E. [US/US]; 418 Huckleberry Drive, Lake Jackson, TX 77566 (US). MANG, Michael, N. [US/US]; 6109 Pinewood Drive, Midland, MI 48640 (US). ZHAO, Jin [CN/US]; 513 Stillmeadow Lane, Midland, MI 48642 (US). (74) Agent: DAMOCLES, Nemia, C.; Patent Dept., P.O. Box 1967, Midland, MI 48641-1967 (US).		(81) Designated States: BR, CA, CN, CZ, JP, KR, MX, PL, RU, US, European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i>
(54) Title: POLYESTER BLENDS (57) Abstract <p>Blends of a hydroxy-functional polyester and a polyalkylene oxide, a polyester polyol or an aliphatic polyester having increased moisture sensitivity and decreased break stress and modulus compared with unblended hydroxy-functional polyesters and are prepared by conventional dry blending methods using conventional means such as a barrel mixer or tumble mixer or by melt blending in an appropriate apparatus, such as a Banbury typed internal mixer. The blends can be used as hot-melt adhesives, adhesive tackifiers, plasticizers, heat-curable adhesives and heat-curable coatings.</p>		

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POLYESTER BLENDS

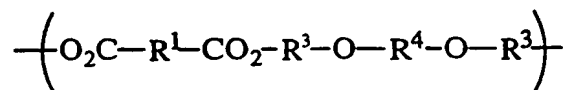
This invention relates to polyester blends, more particularly to blends of hydroxy-functional polyesters, and to processes for preparing the same.

Hydroxy-functional polyesters are known in the art and are described, for example, in U.S. Patents 5,496,910; 5,171,820 and 5,138,022. Blends of hydroxy-functional polyesters with other thermoplastic polyesters are also known, as described in Japanese Patents Shotsugan Kokai 62-25151 and 62-15255. These are immiscible blends which may require compatibilizers to maximize adhesion between phases to insure acceptable physical properties, as described in Japanese Patents Shotsugan Kokai 64-210454. Miscible blends of hydroxy-functional polyesters with thermoplastic polyesters, such as poly(ethylene terephthalate) (PET) are described in U.S. Patent 5,134,201. However, these hydroxy-functional polyesters and blends thereof are not suitable for certain applications, such as hot-melt adhesives, adhesive tackifiers, plasticizers, heat-curable adhesives and heat-curable coatings.

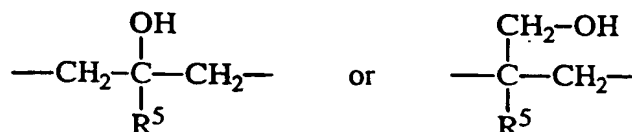
In one aspect, the present invention is a composition comprising a blend of a hydroxy-functional polyester and a polyalkylene oxide, a polyester polyol or an aliphatic polyester.

The blends of the present invention can be used as hot-melt adhesives, adhesive tackifiers, plasticizers, heat-curable adhesives and heat-curable coatings. Some of these materials are biodegradable and are therefore suitable for application to compostable end-products.

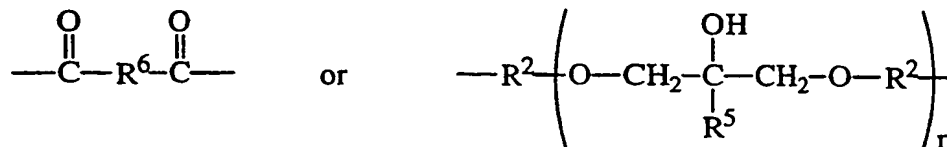
The hydroxy-functional polyester which can be employed in the practice of the present invention is a poly(hydroxy ester) or a poly(hydroxy ester ether) having repeating units represented by the formula:



wherein R^1 is a divalent organic moiety which is primarily hydrocarbon; R^3 is:



and R⁴ is:

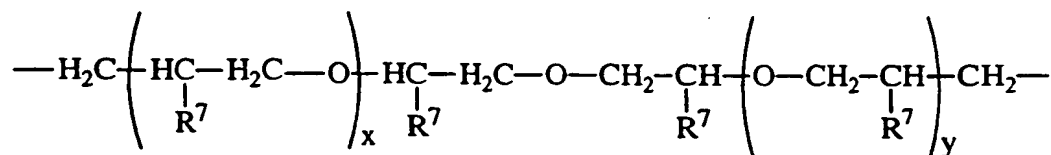


wherein R² and R⁶ are independently divalent organic moieties which are primarily hydrocarbon; R⁵ is hydrogen or alkyl and n is from 0 to 100.

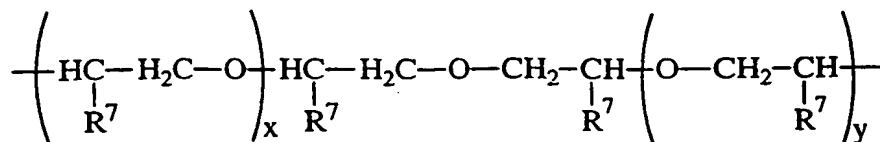
In the preferred polymers, R¹, R² and R⁶ are independently alkylene, cycloalkylene, alkylenearylene, alkyleneoxyalkylene, poly(alkyleneoxyalkylene), alkyleneamidealkylene, poly(alkyleneamidealkylene), alkyleneethioalkylene, poly(alkyleneethioalkylene), alkylene sulfonylalkylene, poly(alkylene sulfonylalkylene), arylene, dialkylenearylene, diaryleneketone, diarylenesulfone, diarylene oxide, alkylidene-diarylene, diarylene sulfide, or a combination of these moieties, optionally substituted with at least one hydroxyl group.

In the more preferred polymers, R¹ is methylene, ethylene, propylene, butylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, nonamethylene, decamethylene, dodecamethylene, 1,4-cyclohexylene, 1,3-cyclohexylene or 1,2-cyclohexylene, optionally substituted with at least one hydroxyl group; and R² and R⁶ are independently methylene, ethylene, propylene, butylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, nonamethylene, decamethylene, dodecamethylene, 1,4-cyclohexylene, 1,3-cyclohexylene or 1,2-cyclohexylene, optionally substituted with at least one hydroxyl group.

More preferably, R¹ and R⁶ are represented by the formula:



and R² is represented by the formula:



wherein R⁷ is independently hydrogen or methyl and x and y are independently 0 to 100.

In the most preferred polymers, R¹ and R⁶ are independently m-phenylene, p-phenylene or 2,6-naphthalene; R² is independently m-phenylene, p-phenylene, naphthalene, diphenylene-isopropylidene, sulfonyldiphenylene, carbonyldiphenylene, oxydiphenylene or 9,9-fluorenediphenylene; R⁵ is hydrogen; R⁷ is independently hydrogen or methyl.

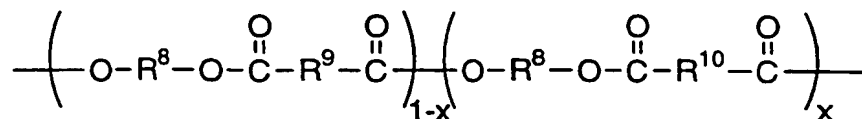
Generally, the hydroxy-functional polyesters can be prepared by reacting dicarboxylic acids and diglycidyl ethers or diglycidyl esters at conditions sufficient to yield hydroxy ester ether or hydroxy ester linkages. These polyesters are described in, U.S. Patent 5,171,820 and copending U.S. Patent Application Serial No. (C-42235) filed October 22, 1996.

Polyalkylene oxides which can be used in the practice of the present invention include those polymers comprising polymerized EO units with an average molecular weight of from 100 to 8,000,000, preferably from 400 to 1,000,000 and most preferably from 1,000 to 100,000.

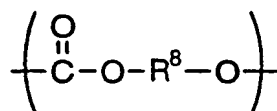
Preferred polyalkylene oxides are poly(ethylene oxide), poly(propylene oxide) poly(ethylene-co-propylene oxide), poly(butylene oxide) and poly(tetrahydrofuran). The most preferred polyalkylene oxide is poly(ethylene oxide).

The polyester polyols which can be used in the practice of the present invention include those prepared by reacting terephthalic acid, isophthalic acid, phthalic anhydride or adipic acid with ethylene glycol. Most preferred is the polyester polyol prepared from adipic acid and ethylene glycol.

The aliphatic polyesters which can be used in the practice of the present invention include polymers having the following repeat units:



or



5

wherein R^8 and R^9 are independently an alkylene moiety such as ethylene, propylene, butylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, methylmethylene, methylethylene, 1-methylpropylene, 2-methylpropylene, 2,2-dimethylpropylene, ethylmethylene, or ethylethylene; R^{10} is an arylene moiety such as 1,4-phenylene, 1,3-phenylene, 1,2-phenylene, 4,4-biphenylene or 2,6-naphthalene; and $x = 0$ to 0.99. Preferred aliphatic polyesters are polycaprolactone, poly(lactic acid), poly(hydroxybutyrate), poly(hydroxybutyrate valerate), poly(butylene succinate) and poly(butylene adipate). The hydroxy-functional polyesters are blended with the polyalkylene oxides, polyester polyol or aliphatic polyester by conventional dry blending methods using conventional means such as a barrel mixer, or a tumble mixer or by melt blending in an appropriate apparatus, such as a Banbury type internal mixer, rubber mill, single or twin screw extruder or compounder.

The amount of the polyalkylene oxide, polyester polyol or aliphatic polyester most advantageously blended with the hydroxy-functionalized polyether is dependent on a variety of factors including the specific polymers used in making the blends, as well as the desired properties of the products resulting from the blends. Typical amounts can range from 1 to 95 weight percent of the blend. Preferably, the polyalkylene oxide, polyester polyol or aliphatic polyester is used in an amount of from 5 to 70 weight percent, more preferably from 10 to 50 weight percent and, most preferably, from 15 to 30 weight percent of the blend.

The following examples are for illustrative purposes only and are not intended to limit the scope of this invention. Unless otherwise indicated, all parts and percentages are by weight.

Examples 1-6

5 A poly(hydroxy ester ether) (PHEE) was synthesized by allowing adipic acid to polymerize with bisphenol A-diglycidyl ether according to the method described in U.S. Patent 5,171,820. In Example 1, a blend of the PHEE (45 g) and poly(ethylene oxide) (PEO, 5 g, MW = 4500) was prepared by mixing the two materials for 15 minutes in a Haake bowl mixer maintained at 120°C to 130°C and with a blade speed of 30 rpm. Other blends
10 (Examples 2-6) of PHEE with PEO having MW = 4500 or 8000 were prepared identically and their compositions are listed in Table I with components expressed as weight percent. The blends were compression molded between sheets made of tetrafluoroethylene fluorocarbon polymers in a 5 by 9 by 0.35 cm mold cavity at a temperature of 80°C and pressure of 100 psi, and the resulting plaques were cut into standard tensile specimens and their mechanical
15 properties determined. The results are shown in Table II.

Comparative Example A

Unblended PHEE was molded and tested as in Examples 1-6. The test results are shown in Table II (Example A).

20 Moldings of the blends in Examples 1-6 and Comparative Example A were immersed in water at 25°C for 350 hours, and water uptake into the samples was determined as percent weight gain (Table II). In addition, films of the blends were cast from solutions in tetrahydrofuran onto glass plates. Contact angles of water droplets placed on the films were measured and are listed in Table II. Increasing wettability of the films are as indicated by decreasing contact angle.

25 Mechanical properties, water uptake and water contact angle of PHEE were determined using the following test procedures:

TEST PROCEDURES**Water Uptake**

The resin was pressed into a 1/8 inch (thickness) by 1 inch (diameter) circular disk mold at 80°C. The circular disk resin was cooled down to room temperature and released from the mold and then vacuum dried overnight at 25°C. The vacuum-dried resin disk was weighed and the weight (W1) recorded. The disk was then placed into a 2 ounce glass bottle filled with 0.3 weight percent of sodium azide in deionized water solution. After 2 weeks immersion in the aqueous solution, the disk was weighed and the weight (W2) recorded. The water uptake was calculated as the percentage of weight gain over the initial dry weight.

$$\text{Water uptake (\%)} = 100 \times (W2 - W1)/(W1)$$

Break Stress: ASTM D3039

Elongation: ASTM D3039

Modulus: ASTM D3039

15 **Contact Angle:** ASTM 971

TABLE I

<u>Example</u>	<u>PHEE (%)</u>	<u>PEO</u> <u>(MW = 4500, %)</u>	<u>PEO</u> <u>(MW = 8000, %)</u>
1	90	10	0
2	80	20	0
3	70	30	0
4	90	0	10
5	80	0	20
6	70	0	30
A	100	0	0

TABLE II

<u>Example</u>	<u>Break Stress</u>	<u>Elongation (%)</u>	<u>Modulus (psi)</u>	<u>Water Uptake (%)</u>	<u>Contact Angle (%)</u>
1	900	363	298	15	81
2	29	778	103	28	--
3	946	31	55,633	37	20
4	738	333	626	14	69
5	13	1215	103	33	72
6	1661	4	75,429	34	--
A	2900	25	402,000	3	90

The dramatic decrease in break stress and modulus in blends of PHEE and PEO, compared
 with those of PHEE alone, as shown in the above Table II, indicated that PEO can plasticize
 PHEE owing to the surprising miscibility or partial miscibility of poly(alkylene oxides) in
 poly(hydroxy ester ethers). Also, incorporation of PEO in PHEE lead to substantially
 increased moisture sensitivity in the blends compared with unmodified PHEE, reflected by
 high water uptake (14 to 37 percent) and low water contact angles (20 to 81 percent) of
 Examples 1-6. Alone, PHEE absorbed only about 3 percent moisture and cast films had a
 high water contact angle of 90 percent. The ability of poly(alkylene oxides) to plasticize
 biodegradable poly(hydroxy ester ethers) and the capability of PEO to increase the moisture
 sensitivity of the polymers indicate that PHEE/poly(alkylene oxide) blends are useful for hot-
 melt adhesives intended to disintegrate in water and biodegrade during post-consumer
 disposal.

Example 7

Binary mixtures of the poly(hydroxy ester ether) derived from bisphenol A diglycidyl ether and 1,4-cyclohexanedicarboxylic acid and polycaprolactone, poly(lactic acid) or a commercial blend of polycaprolactone and starch (Bioplast, produced by Biotec Naturverpackungen GmbH) were prepared by mixing in the melt state in a Brabender roller mixer. First, the blend of desired amounts of two polymers was quickly fed into the mixing bowl of the mixer that was preheated to the desired temperature (180°C for blends with polycaprolactone; 200°C for blends with poly(lactic acid); and 160°C for the blends with the commercial Bioplast resin). During this feeding step, the mixing blades were set at a low speed, but after the chamber was completely full, the lid was closed and the speed increased. Upon addition of the charge, the temperature of the chamber initially decreased by 10°C to 20°C, but it regained its original level in 4 to 5 minutes. The melt-blended mixtures were then compression molded between tetrafluoroethylene-coated plates to form films or rectangular bars for tensile and dynamic mechanical testing. All of the blend samples were also conditioned in the constant temperature room (50 percent relative humidity and 23°C) for at least 24 hours before they were tested. The results of the tests are shown in Tables III and IV.

Table III

Tensile properties of a binary blend of the poly(hydroxy ester ether) derived from bisphenol A diglycidyl ether and 1,4-cyclohexanedicarboxylic acid with polycaprolactone measured on compression molded test specimens.

Weight Percentage Polycaprolactone in Blend	Tensile Elongation (%)	True Stress at Break (MPa)	Tensile Modulus (MPa)
0	41	51	1944
25	374	107	83
50	627	268	461
75	732	333	390
100	987	658	295

Table IV

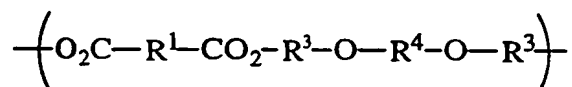
Tensile properties of a binary blend of the poly(hydroxy ester ether) derived from bisphenol A diglycidyl ether and 1,4-cyclohexanedicarboxylic acid with a blend of polycaprolactone and starch measured on compression molded test specimens.

Weight Percentage of PHEE in Blend	Tensile Elongation (%)	True Stress to Break (MPa)
0	628	89
5	891	166
10	792	158
20	788	182

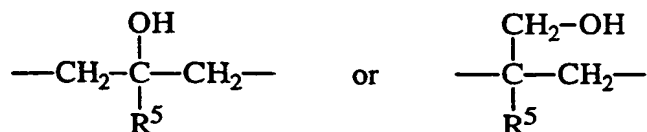
CLAIMS:

1. A composition comprising a blend of a hydroxy-functionalized polyester and a polyalkylene oxide, a polyester polyol or an aliphatic polyester.

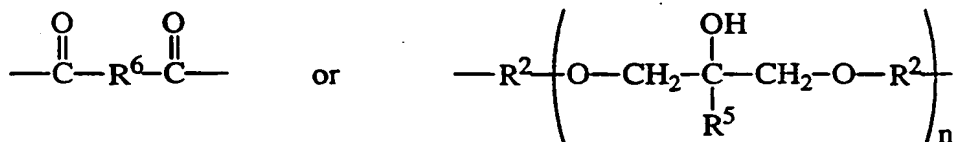
2. The composition of Claim 1 wherein the hydroxy-functionalized polyester is
 5 a poly(hydroxy ester) or a poly(hydroxy ester ether) having repeating units represented by the formula:



wherein R¹ is a divalent organic moiety which is primarily hydrocarbon; R³ is:



10 and R⁴ is:

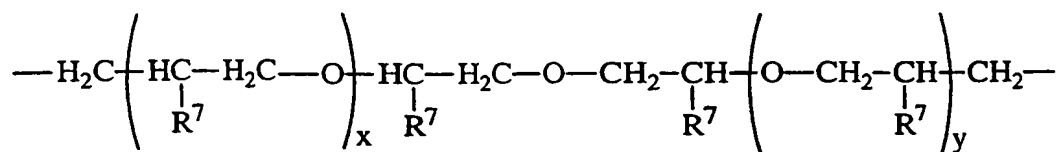


wherein R² and R⁶ are independently divalent organic moieties which are primarily hydrocarbon; R⁵ is hydrogen or alkyl and n is from 0 to 100.

3. The composition of Claim 2 wherein R¹ is independently alkylene,
 15 cycloalkylene, alkylenearylene, alkyleneoxyalkylene, poly(alkyleneoxyalkylene),
 alkyleneethioalkylene, alkylene sulfonylalkylene, optionally substituted with at least one
 hydroxy group; arylen, dialkylenearylene, diaryleneketone, diarylenesulfone, diarylene
 oxide, alkylidene-diarylene, diarylene sulfide, or a combination of these moieties.

4. The composition of Claim 2 wherein R¹ is independently methylene,
 20 ethylene, propylene, butylene, pentamethylene, hexamethylene, heptamethylene,
 octamethylene, nonamethylene, decamethylene, dodecamethylene, 1,4-cyclohexylene,
 1,3-cyclohexylene or 1,2-cyclohexylene, optionally substituted with at least one hydroxyl
 group.

5. The composition of Claim 2 wherein R¹ is represented by the formula:



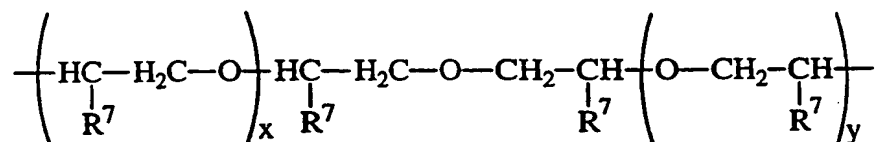
wherein R⁷ is independently hydrogen or methyl and x and y are independently 0 to 100.

6. The composition of Claim 2 wherein R¹ is independently m-phenylene, p-phenylene or 2,6-naphthalene.

7. The composition of Claim 2 wherein R² is independently alkylene, cycloalkylene, alkylenearylene, alkyleneoxyalkylene, poly(alkyleneoxyalkylene), alkyleneethioalkylene, alkylene sulfonylalkylene, optionally substituted with at least one hydroxy group; arylene, dialkylenearylene, diaryleneketone, diarylenesulfone, diarylene oxide, alkylidene-diarylene, diarylene sulfide, or a combination of these moieties.

8. The composition of Claim 2 wherein R² is independently ethylene, propylene, butylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, nonamethylene, decamethylene, dodecamethylene, 1,4-cyclohexylene, 1,3-cyclohexylene or 1,2-cyclohexylene, optionally substituted with at least one hydroxyl group.

9. The composition of Claim 2 wherein R² is represented by the formula:



wherein R⁷ is independently hydrogen or methyl and x and y are independently 0 to 100 and n is O.

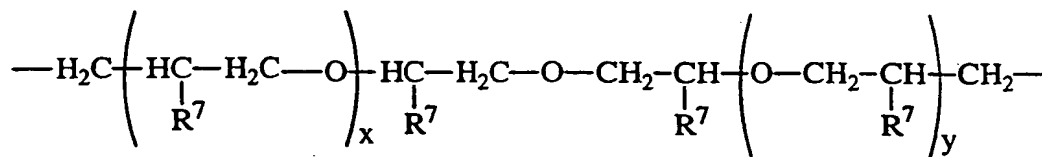
10. The composition of Claim 2 wherein R² is m-phenylene, p-phenylene, naphthalene, diphenylene-isopropylidene, sulfonyldiphenylene, carbonyldiphenylene, oxydiphenylene or 9,9-fluorenediphenylene.

11. The composition of Claim 2 wherein R⁶ is independently alkylene, cycloalkylene, alkylenearylene, alkyleneoxyalkylene, poly(alkyleneoxyalkylene), alkyleneethioalkylene, alkylene sulfonylalkylene, optionally substituted with at least one

hydroxy group; arylene, dialkylenearylene, diaryleneketone, diarylenesulfone, diarylene oxide, alkylidene-diarylene, diarylene sulfide, or a combination of these moieties.

12. The composition of Claim 2 wherein R⁶ is independently ethylene, propylene, butylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, nonamethylene, decamethylene, dodecamethylene, 1,4-cyclohexylene, 1,3-cyclohexylene or 1,2-cyclohexylene, optionally substituted with at least one hydroxyl group.

13. The composition of Claim 2 wherein R⁶ is represented by the formula:



wherein R⁷ is independently hydrogen or methyl and x and y are independently 0 to 100.

14. The composition of Claim 2 wherein the hydroxy-functionalized polyester is a poly(hydroxy ester ether) in which R¹ is butylene, R² is diphenylene-isopropylidene, R⁵ is H, and n is O.

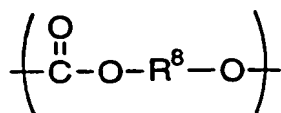
15. The composition of Claim 1 wherein the polyalkylene oxide is poly(ethylene oxide).

16. The composition of Claim 1 wherein the polyalkylene oxide is poly(propylene oxide).

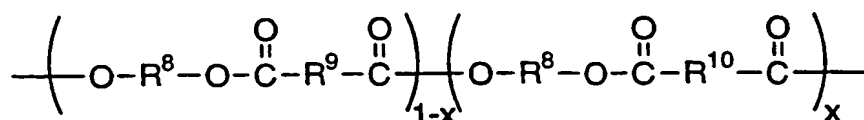
17. The composition of Claim 1 wherein the polyalkylene oxide is poly(ethylene-co-propylene oxide).

18. The composition of Claim 1 wherein the polyester polyol is formed by the reaction of terephthalic acid, isophthalic acid, phthalic anhydride or adipic acid and ethylene glycol.

19. The composition of Claim 1 wherein the aliphatic polyester is represented by the formulae:



or



- 5 wherein R^8 and R^9 are independently an alkylene moiety, R^{10} is an arylene moiety and $x = 0$ to 0.99.

20. The composition of Claim 1 wherein the aliphatic polyester is polycaprolactone, poly(lactic acid), poly(hydroxybutyrate), poly(hydroxybutyrate valerate), poly(butylene succinate) or poly(butylene adipate).

- 10 21. The composition of Claim 1 wherein the hydroxy-functionalized polyester is present in an amount of from 5 to 99 weight percent, based on the weight of the blend.

22. The composition of Claim 1 prepared by mixing a poly(hydroxy ester) or a poly(hydroxy ester ether) and a polyalkylene oxide, a polyester polyol or an aliphatic polyester in a molten state.

- 15 23. A hot-melt adhesive comprising the composition of Claim 1.

24. An adhesive tackifier comprising the composition of Claim 1.

25. A plasticizer comprising the composition of Claim 1.

26. The composition of Claim 1 in the form of a film, a flexible or rigid foam, molded article, fiber, coating or laminate.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 97/22892

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08L67/00 C09J167/00

According to International Patent Classification(IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08L C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 741 178 A (NATIONAL STARCH AND CHEMICAL INVESTMENT) 6 November 1996 see claims 1-3,8-10	1-15, 19-25
X	EP 0 741 177 A (NATIONAL STARCH AND CHEMICAL INVESTMENT) 6 November 1996 see claims 1,4	1-19, 21-25

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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